

FORM PTO-1300  
(REV. 12-2001)

U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NUMBER

MPL318

TRANSMITTAL LETTER TO THE UNITED STATES  
DESIGNATED/ELECTED OFFICE (DO/EO/US)  
CONCERNING A FILING UNDER 35 U.S.C. 371

U.S. APPLICATION NO. (If known, see 37 CFR 1.5

10/069090

INTERNATIONAL APPLICATION NO.  
PCT/GB00/02725INTERNATIONAL FILING DATE  
14 July 2000PRIORITY DATE CLAIMED  
17 July 1999

TITLE OF INVENTION

STRUCTURED SURFACTANT SYSTEMS

APPLICANT(S) FOR DO/EO/US  
HAWKINS, John

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
  - a. ☒ is attached hereto (required only if not communicated by the International Bureau).
  - b. ☐ has been communicated by the International Bureau.
  - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☐ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
  - a. ☐ is attached hereto.
  - b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
  - a. ☐ are attached hereto (required only if not communicated by the International Bureau).
  - b. ☐ have been communicated by the International Bureau.
  - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
  - d. ☐ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. ☐ An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).

## Items 11 to 20 below concern document(s) or information included:

11. ☒ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. ☒ A **FIRST** preliminary amendment.
14. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
15. ☐ A substitute specification.
16. ☒ A change of power of attorney and/or address letter.
17. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
18. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
19. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
20. ☐ Other items or information:

CERTIFICATE OF MAILING BY EXPRESS MAIL  
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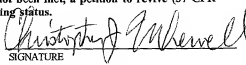
page 1 of 2

I hereby certify that the application and correspondence attached hereto is being deposited with the U.S. Postal Service "Express Mail Post Office to Addressee" Service under 37 CFR 1.10 on the date indicated below and is addressed to Commissioner for Patents, Washington, D.C. 20231

*Martha Victory*  
Martha Victory

*Feb. 7, 2002*  
Date of Deposit

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U.S. APPLICATION NO. (if known, see 37 CFR 1.51) <b>10/069090</b>		INTERNATIONAL APPLICATION NO. PCT/GB00/02725		ATTORNEY'S DOCKET NUMBER MFL318	
21. <input type="checkbox"/> The following fees are submitted: <b>BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)):</b> Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO ..... <b>\$1040.00</b> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO ..... <b>\$890.00</b> International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO ..... <b>\$740.00</b> International preliminary-examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) ..... <b>\$710.00</b> International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) ..... <b>\$100.00</b> <b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>				<b>CALCULATIONS PTO USE ONLY</b>	
				\$ 890.00	
Surcharge of \$130.00 for furnishing the oath or declaration later than months from the earliest claimed priority date (37 CFR 1.492(e)). <input type="checkbox"/> 20 <input checked="" type="checkbox"/> 30				\$ 130.00	
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE	\$	
Total claims	- 20 =		x \$18.00	\$	
Independent claims	4 - 3 =	1	x \$84.00	\$ 84.00	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)			+ \$280.00	\$	
<b>TOTAL OF ABOVE CALCULATIONS =</b>				<b>\$ 1104.00</b>	
<input type="checkbox"/> Applicant claims small entity status. See 37 CFR 1.27. The fees indicated above are reduced by 1/2.				\$	
<b>SUBTOTAL =</b>				<b>\$ 1104.00</b>	
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$	
<b>TOTAL NATIONAL FEE =</b>				<b>\$ 1104.00</b>	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property +				<b>\$ 40.00</b>	
<b>TOTAL FEES ENCLOSED =</b>				<b>\$ 1144.00</b>	
				Amount to be refunded:	\$
				charged:	<b>\$ 1144.00</b>
a. <input type="checkbox"/> A check in the amount of \$ _____ to cover the above fees is enclosed.					
b. <input checked="" type="checkbox"/> Please charge my Deposit Account No. <u>08-3442</u> in the amount of \$ <u>1144.00</u> to cover the above fees. A duplicate copy of this sheet is enclosed.					
c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>08-3442</u> . A duplicate copy of this sheet is enclosed.					
d. <input type="checkbox"/> Fees are to be charged to a credit card. <b>WARNING:</b> Information on this form may become public. <b>Credit card information should not be included on this form.</b> Provide credit card information and authorization on PTO-2038.					
NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.					
SEND ALL CORRESPONDENCE TO:					
Huntsman Corporation Legal Department P. O. Box 15730 Austin, TX 78761			SIGNATURE  Christopher J. Whewell NAME 37,469 REGISTRATION NUMBER		

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants:	John Hawkins	§	
		§	Docket # MPL318
		§	
Serial No. unk. (entry into National Stage in US		§	
of PCT/GB00/02725		§	
		§	
Filed: concurrently herewith		§	
		§	
For: "STRUCTURED SURFACTANT		§	
SYSTEMS"		§	

January 23, 2002

Commissioner for Patents  
 Washington, D.C. 20231

Preliminary Amendment

Sir:

Prior to calculation of the filing fee in the above-captioned matter, please enter the following amendments to the claims:

1) (AMENDED) A structured surfactant system comprising a water soluble carbohydrate as a structurant.

5) (AMENDED) A system according to claim 4 which is transparent.

8) (AMENDED) A system according to claim 2 wherein said carbohydrate is present in an amount of at least 25 % by weight based on the weight of said system.

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9) (AMENDED) A system according to claim 2 wherein the surfactant is present in any amount between 1 to 60% by weight based on the weight of the system.

10) (AMENDED) A system according to claim 2 wherein water is present in an amount greater than 20 % by weight based on the weight of the system.

12) (AMENDED) A system according to claim 2 wherein said carbohydrate is selected from the group consisting of: mono saccharides sugars, disaccharide sugars, gluconic acid, mannitol, ascorbic acid, sorbitol, mannitol or inositol.

13) (AMENDED) An aqueous suspension comprising a system according to claim 2 and further comprising particles of solid, liquid or gas stably suspended therein.

15) (AMENDED) A suspension according to claim 13 wherein said particles comprise an abrasive.

16) (AMENDED) A suspension according to claim 13 wherein said particles comprise a pesticide.

17) (AMENDED) A suspension according to claim 13 wherein said particles comprise an oil.

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18) (AMENDED) A suspension according to claim 13 wherein said particles comprise a pigment.

Claims 1, 5, 8, 9, 10, 12, 13, 15, 16, 17, and 18 have been amended herein.

Attached hereto is a marked-up version of the changes made to the claims by the current amendment. The attached page is captioned: "Version with markings to show changes made".

Respectfully Submitted,



Christopher J. Whewell, Reg. No. 37,469  
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Version with markings to show changes made

1) (AMENDED) ~~The use of water soluble surfactants as structurants in structured surfactant systems.~~ A structured surfactant system comprising a water soluble carbohydrate as a structurant.

5) (AMENDED) A ~~composition system~~ according to claim 4 which is transparent.

8) (AMENDED) A system according to ~~any of claims 2 to 7~~ wherein said carbohydrate is present in ~~any an~~ amount of at least 25 % by weight ~~of the mixture of water surfactant-carbohydrate~~ based on the weight of said system.

9) (AMENDED) A system according to ~~any of claims 2 to 8~~ wherein the surfactant is present in ~~an any~~ amount ~~of from~~ between 1 to 60% by weight based on the weight of the system.

10) (AMENDED) A system according to ~~any of claims 2 to 9 containing more~~ wherein water is present in an amount greater than 20 % by weight ~~water~~ based on the weight of the system.

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12) (AMENDED) A ~~composition-~~ system according to ~~any of~~ claims 2 to 11 wherein ~~the said~~ carbohydrate is selected from the group consisting of: -a mono saccharides sugars, or disaccharide sugars, gluconic acid, mannitol, ascorbic acid, sorbitol, mannitol or inositol.

13) (AMENDED) An aqueous suspension comprising a system according to ~~any of~~ claims 2 to 7 and further comprising particles of solid, liquid or gas stably suspended therein.

15) (AMENDED) A ~~composition-~~ suspension according to ~~either of~~ claims 13 and 14 wherein said particles comprise an abrasive.

16) (AMENDED) A ~~composition-~~ suspension according to ~~any of~~ claims 13 to 16 wherein said particles comprise a pesticide.

17) (AMENDED) A ~~composition-~~ suspension according to ~~any of~~ claims 13 to 16 wherein said particles comprise an oil.

18) (AMENDED) A ~~composition-~~ suspension according to ~~any of~~ claims 13 to 17 wherein said particles comprise a pigment.

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## STRUCTURED SURFACTANT SYSTEMS

The present invention relates to the formulation of structured surfactant suspending systems. It is particularly relevant to the formulation of personal care formulations such as shampoos and skin cleansing preparations.

### STRUCTURED SURFACTANT

Suspending solids in liquids presents a problem. If the solids differ in density from the liquid they will tend either to sediment or float. Increasing the viscosity of the liquid can retard, but not prevent such separation, and high viscosities are generally undesirable. Colloidal systems, in which the suspended particles are sufficiently small to experience Brownian motion, e.g. less than 1 micron, may be kinetically stable. However the difficulty or undesirability of comminuting some solids to such sizes, and the impossibility of maintaining many of them at this level in the face of crystal growth or agglomeration, limits the use of colloidal suspensions.

Adjusting the density of one phase to match that of the other is usually impracticable. Moreover such systems are almost always temperature-unstable due to differential rates of thermal expansion.

One method of suspension which permits even relatively large particles to be stably suspended is structured surfactant. The term covers systems in which a surfactant mesophase, usually a lamellar or G-phase, alone or more usually interspersed with an aqueous phase, provides a yield stress which is sufficient, when the system is at rest, to immobilise any suspended particles, but which is sufficiently low to allow the system to be poured like a normal liquid. Such systems may display very low apparent viscosities when stirred, pumped or poured and yet be capable of maintaining particles, sometimes of millimetre or larger size, indefinitely in suspension.

Three main types of suspending system have been employed in practice, all involving

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a G-phase, in which bilayers of surfactant are arranged with the hydrophobic part of the molecule on the interior and the hydrophilic part on the exterior of the bilayer (or vice versa). The bilayers lie side by side, e.g. in a parallel or concentric configuration, sometimes separated by aqueous layers. G-phases (also known as  $L_{\alpha}$  phases) can usually be identified by their characteristic textures under the polarising microscope and/or by x-ray diffraction, which is often able to detect evidence of lamellar symmetry. Such evidence may comprise first, second and sometimes third order peaks with d-spacing ( $\frac{2\pi}{Q}$  where  $Q$  is the momentum transfer vector) in a simple integral ratio 1:2:3. Other types of symmetry give different ratios, usually non-integral.

The d-spacing of the first peak in the series corresponds to the repeat spacing of the bilayer system.

Most surfactants form a G-phase either at ambient or at some higher temperature when mixed with water in certain specific proportions. However such conventional G-phases do not usually function as structured suspending systems. Useful quantities of solid render them unpourable and smaller amounts tend to sediment.

The main types of structured system used in practice are based on dispersed lamellar, spherulitic and expanded lamellar phases. Dispersed lamellar phases are two phase systems in which the surfactant bilayers are arranged as parallel plates to form domains of G-phases which are interspersed with an aqueous phase to form an opaque gel-like system. They are described in EP O 086 614.

Spherulitic phases comprise well defined spheroidal bodies, usually referred to in the art as spherulites, in which surfactant bilayers are arranged as concentric shells. The spherulites usually have a diameter in the range 0.1 to 15 microns and are dispersed in an aqueous phase in the manner of a classical emulsion, but interacting to form a structured system. Spherulitic systems are described in more detail in EP O 151 884.

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Many structured surfactant systems are intermediate between dispersed lamellar and spherulitic, involving both types of structure. Usually systems having a more spherulitic character are preferred because they tend to have lower viscosity. A variant on the spherulitic system comprises prolate or rod shaped bodies sometimes referred to as batonnettes.

Both of the foregoing systems comprise two phases. Their stability depends on the presence of sufficient dispersed phase to pack the system so that the interaction between the spherulites or other dispersed mesophase domains prevents separation. If the amount of dispersed phase is insufficient, e.g. because there is not enough surfactant or because the surfactant is too soluble in the aqueous phase to form sufficient of a mesophase, the system will undergo separation and cannot be used to suspend solids. Such unstable systems are not considered to be "structured" for the purpose of this specification.

A third type of structured surfactant system comprises an expanded G-phase. It differs from the other two types of structured system in being essentially a single phase, and from conventional G-phase in having a wider d-spacing. Conventional G-phases, which typically contain 60 to 75% by weight surfactant, have a d-spacing of about 4 to 7 nanometers. Attempts to suspend solids in such phases results in stiff pastes which are either non-pourable, unstable or both. Expanded G-phases with d-spacing greater than 8, e.g. 10 to 15 nanometers, form when the electrolyte is added to aqueous surfactants at concentrations just below those required to form a normal G-phase, particularly to surfactants in the M phase. The M phase comprises surfactant molecules arranged to form cylindrical rods of indefinite length. It exhibits hexagonal symmetry and a distinctive texture under the polarising microscope. Typical M phases have so high a viscosity that they appear to be curdy solids. M phases near the lower concentration limit (the  $L_1/M$  phase boundary) may be pourable but have a very high viscosity and often a mucous-like appearance. Such systems tend to form expanded G-phases particularly readily on addition of sufficient electrolyte.

Expanded G-phases are described in more detail in EP O 530 708. In the absence of suspended matter they are translucent, unlike dispersed lamellar or spherulitic phases which are necessarily opaque. They are optically anisotropic and have shear dependent viscosity. In this they differ from  $L_1$  phases which are micellar solutions and which include microemulsions.  $L_1$  phases are clear, optically isotropic and are usually substantially Newtonian. They are unstructured and cannot suspend solids.

Some  $L_1$  phases exhibit small angle x-ray diffraction spectra which show evidence of hexagonal symmetry and/or exhibit shear dependent viscosity. Such phases usually have concentrations near the  $L_1/M$  phase boundary and may form expanded G-phases on addition of electrolyte. However in the absence of any such addition of electrolyte they lack the yield point required to provide suspending properties, and are therefore not considered to be "structured systems" for the purpose of this specification.

Expanded G phases are usually less robust than spherulitic systems. They are liable to undergo a phase change at elevated temperatures to the optically-isotropic, unstructured  $L_2$  phase. Relatively low yield stress may limit the maximum size of particle that can be stably suspended.

Most structured surfactants require the presence of electrolyte as well as surfactant and water in order to form structured systems capable of suspending solids. However certain relatively hydrophobic surfactants such as isopropylamine alkyl benzene sulphonate can form spherulites in water in the absence of electrolyte. Such surfactants are capable of suspending solids in the absence of electrolyte as described in EP O 414 549.

#### **APPLICATION**

Structured surfactants have been applied to the problems of suspending: water insoluble or sparingly soluble builders in laundry detergent; antifoams and enzymes in laundry detergents and other surfactant systems; abrasives in hard surface cleaners; pesticides and oils in agrochemical preparations (EP O 388 239 and EP O 498 231);

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rock cuttings in drilling muds (EP O 430 602); dyestuffs in dyebath concentrates and printing inks (EP O 472 089); talcs, oils and other cosmetic ingredients in personal care formulations (EP O 530 708). The present invention is applicable to all the foregoing. It is especially applicable to cosmetic and personal care formulations in which the physical appearance of the product may be a major factor in promoting sales, for example, to shampoos, body lotions, shower gels or hair creams. It may also be applied to pharmaceutical preparations such as, drug delivery systems, and to flavourings and other concentrates for the food industry and to toothpastes.

### **FLOCCULATION**

A problem with the two phase structured surfactant systems, and especially spherulitic systems, is flocculation of the dispersed surfactant structures. This tends to occur at high surfactant and/or high electrolyte concentration. It can have the effect of making the composition very viscous and/or unstable with the dispersed surfactant separating from the aqueous phase.

Certain amphiphilic polymers have been found to act as deflocculants of structured surfactants. One type of deflocculant polymer exhibits cteniform (comb-shaped) architecture with a hydrophilic backbone and hydrophobic side chains or vice versa. A typical example is a random copolymer of acrylic acid and a fatty alkyl acrylate. Cteniform deflocculants have been described in a large number of patents, for example WO-A-9106622.

A more effective type of deflocculant has surfactant rather than cteniform architecture, with a hydrophilic polymer group attached at one end to a hydrophobic group. Such deflocculants are typically telomers formed by telomerising a hydrophilic monomer with a hydrophobic telogen. Examples of surfactant deflocculants include alkyl thiol polyacrylates and alkyl polyglycosides. Surfactant deflocculants are described in more details in EP O 623 670.

In a copending PCT patent application PCT/GB00/02447 filed on 22 June 2000 claiming priority from British patent application no. 9914673 we have described the use of small amounts (e.g. about 15% by weight of the composition) of carbohydrates such as sugars and alginates as deflocculants in structured surfactant compositions. The latter comprise surfactant, water and electrolyte in proportions adapted to form flocculated two-phase structured surfactant systems in the absence of the carbohydrate.

### **THE PROBLEM**

Existing structured surfactant formulations are constrained by several limitations which have hitherto limited their application, especially in the areas of cosmetics and personal care. These include the following:-

1. Unless a substantial amount of electrolyte is present the choice of surfactant is limited to a fairly small range of relatively insoluble surfactants such as isopropyl alkyl benzene sulphonates. For many applications these are not the surfactants of choice from a performance point of view, and in some cases are totally inappropriate.
2. Spherulitic or dispersed lamellar structured surfactants are opaque. This limits the visual effects that can be achieved and may be perceived as less attractive than a clear system in some applications.
3. Expanded G phases are normally opalescent, have limited suspending power and are usually formed over narrow concentrations and/or temperature ranges which make them difficult to use in practice.
4. At high surfactant concentrations, e.g. above 25% by weight it is difficult to make stable structured systems without using expensive deflocculants and auxiliary stabilisers.

5. Most surfactant systems require preservatives to prevent microbial spoilage. However preservatives are expensive, ecologically undesirable, and may cause sensitivity problems for some users.

There is a need, especially in the personal care field, for a suspending system that is clear, transparent and mobile. There is a need for a system which contains high levels of surfactant but which does not require expensive deflocculants. There is a need for a system that contains relatively soluble surfactants but which does not require the presence of electrolyte as a structurant. There is a need for a cleaning composition which does not require added preservatives.

### THE SOLUTION

We have now discovered that formulations meeting some or all of the above needs, may be obtained by using water soluble carbohydrate to impart structure to the surfactant system, instead of or in addition to the electrolytes used hitherto. A component of the structured surfactant system which is used to impart structure to the surfactant will be referred to herein as a "structurant".

### THE INVENTION

Our invention provides the use of water soluble carbohydrates as structurants in structured surfactant suspending systems.

According to a second embodiment our invention provides a structured surfactant system having suspending properties which comprises a surfactant, water and a structurant characterised in that said structurant comprises a water soluble carbohydrate. The structured system may typically be a very highly expanded G-phase, e.g. one having a lamellar repeat spacing greater than 8 and usually greater than 15nm. The composition, in the absence of suspended matter, is preferably clear and transparent.

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According to a further embodiment the invention provides a structured surfactant system having suspending properties and comprising surfactant, carbohydrate and water and having a structural feature with a repeat spacing of 20 to 50nm. The structural feature is preferably lamellar, e.g. consisting of a highly expanded G-phase.

According to a third embodiment the invention provides a composition comprising a structured surfactant system of the invention as specified above and suspended particles. The particles may be solid, liquid or gaseous and are either insoluble in the composition or present in excess of their solubility.

### **THE STRUCTURED SYSTEM**

The term "structured system" as used herein means a pourable composition comprising water, surfactant, dissolved carbohydrate and any other dissolved matter, including any costructurants, which together form a mesophase, or a dispersion of a mesophase in a continuous aqueous medium, and which has the ability to immobilise suspended particles while the system is at rest, to form a pourable suspension.

The aqueous structured systems formed by the interaction of surfactants with carbohydrates include systems which are believed to be in the form of an expanded G-phase. In particular they include novel systems having a much wider repeat spacing than the typical electrolyte-structured expanded G-phases described in EP O 530 708. The systems of the present invention comprise structures which typically show a repeat spacing between 20 and 50nm which is approximately double the repeat spacings measured for electrolyte-structured expanded G-phase, and approximately four times the typical repeat spacing in a conventional binary surfactant/water G-phase. The following discussion is based on the assumption that the structure is lamellar. We do not, however, intend to exclude the possibility that the system may comprise non-lamellar components.

Surprisingly, despite the apparently high lamellar spacing of the G-phases of the

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present invention, they are generally robust with good suspending power and good temperature stability. Typically the viscosity increases slightly with increasing temperature and the systems are often stable up to 70°C or higher.

The systems when fully deaerated and free from suspended fine insoluble particles are generally obtainable in a substantially clear and transparent form in marked contrast to other structured surfactant systems. This can typically be achieved by vigorous centrifugal deaeration and/or by gentle heating at, e.g. 60 to 80°C.

If the amount of surfactant or of structurant is not sufficiently high, or the ratio of electrolyte to carbohydrate is too high, the structured system of the invention will be obtained as an opaque two phase system which may be spherulitic or comprise dispersed G-phase or batonettes.

### **PROPORTIONS**

The proportions vary depending on the nature of the surfactant and of the carbohydrate. Figure 1 shows, in schematic form, a typical phase diagram for the system coconut diethanolamide, sucrose and water. The area marked "clear lamellar" represents the clear, pourable structured suspending system. The following typical proportions are expressed by weight of the total structured system i.e. comprising the water, surfactant, structurant and any other dissolved matter but excluding any suspended solids or water-immiscible liquids.

Generally the surfactant is present in an amount of at least 2%, e.g. at least 5% especially more than 10%, by weight of the system but preferably less than 60% e.g. less than 50%, especially less than 40% more especially less than 30%. A convenient range is 3 to 25% especially 4 to 12%.

Carbohydrate structurants are usually required in substantially higher proportions than would be required for an electrolyte structurant. Preferably in the absence of electrolyte, the carbohydrate is present in a proportion of at least 25% e.g. at least



30% and usually more than 40% by weight. Concentrations greater than 65% are usually avoided. Typically the carbohydrate is less than 60%, usually less than 55% by weight of the composition. When electrolyte is present the carbohydrate may be present in substantially lower concentrations as a costructurant. Such systems may be spherulitic but in the presence of more than about 10% sugar, do not tend to flocculate. The systems of the invention require the presence of the carbohydrate in order to form a structured suspending system. Typically the less soluble the surfactant, the less carbohydrate is required.

The proportion of water is usually greater than 20% by weight, more commonly greater than 30%, typically greater than 40% of the system, but is preferably less than 65% usually less than 60%, e.g. less than 55%.

One way of preparing suspending systems according to the invention is to prepare a G or M phase aqueous surfactant and add sugar until the system clears. The G or M phases are located using conventional means, as described for example in GB 2 013 235.

Suspending power may be quickly checked by shaking air into the sample and noting whether the bubbles remain suspended. Confirmation that the system is a true structured system and not merely a slowly separating system may be obtained by allowing the sample to stand overnight at 50 or 60°C. If the dispersed phase has not separated out in that time, the system may be assumed to be structured. It is generally found that mixtures of two G-phase systems according to the invention also form G-phases according to the invention.

If in any case difficulty is encountered locating a sugar-structured phase according to the invention, it is usually possible to resolve by adding a minor proportion based on the weight of sugar of a co-structurant has discussed below.

## THE CARBOHYDRATE

The preferred carbohydrates are mono and disaccharide sugars such as sucrose, glucose or fructose. Other sugars which can be used include mannose, ribose, galactose, allose, talose, gulose, idose, arabinose, xylose, lyxose, erythrose, threose, acrose, rhamose and cellobiose. The carbohydrate may be a tri- or tetra-saccharide or a water soluble polysaccharide such as soluble starch. The term "carbohydrate" as used here includes water soluble non-surfactant derivatives of carbohydrates such as carboxylic acids and their salts, e.g. gluconic acid, mannic acid, ascorbic acid and alginates or reduced sugars such as sorbitol, mannitol or inositol. The levels of carbohydrate are preferably sufficiently high to inhibit microbiological growth in the medium and preferably sufficient to act as an effective biodegradable, non-allergenic preservative for the composition, thereby obviating the need for less environmentally friendly additives.

## CO-STRUCTURANT

Some surfactants, especially the more water soluble surfactants such as alkyl ether sulphates form the clear lamellar phase more readily in the presence of a co-structurant. The co-structurant is preferably an electrolyte. Any water soluble salt which tends to lower the solubility of surfactant in water may be used, such as sodium tripolyphosphate, sodium carbonate, sodium citrate, sodium chloride or the corresponding potassium or ammonium salts. Alkalis such as sodium or potassium hydroxide may also be used. Other structurants include polar water-immiscible solvents such as phenoethoxy ether or a terpene. water soluble mono and dihydroxy alcohols and ether alcohols such as glycerol, propylene glycol, ethylene glycol monomethyl ether and diethylene glycol monomethyl ether.

The constructurant, if required, may in principal, be present in concentrations up to 30%, but is preferably less than 20% e.g. 0.1 to 15% by weight. Often traces of costructurants e.g. 0.1 to 3%, typically 0.5 to 2.5% by weight based on the system are sufficient, although higher concentrations can be present. For example in some

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perfumed systems, the solvent in the perfume may be sufficient to provide any desired co-structuring effect. We prefer that the proportion of costructurant be less than the proportion of carbohydrate, preferably less than half the proportion of carbohydrate, e.g. less than one quarter the proportion of carbohydrate. Large amounts of electrolyte are generally undesirable because they inhibit the formation of clear phases.

Generally the costructurant is present in proportions insufficient to form a stable structured system in the absence of the carbohydrate.

### THE SURFACTANT

The surfactant preferably comprises non-ionic surfactants such as  $C_{8-25}$  alkyl mono or diethanolamides or 1 to 50 mole ethoxylates such as  $C_{8-25}$  alcohol or fatty acid ethoxylates, alkyl amine ethoxylates, or glyceryl or sorbitan ester ethoxylates, or polyoxypropylene oxyethylene block copolymers. Ethoxylates typically contain from 2 to 40 eg. 3 to 30 especially 5 to 15 oxyethylene groups. Other non-ionic surfactants include alkyl polyglycosides, sugar esters or amine oxides. The non-ionic surfactants typically have a HLB of from 5 to 16, e.g. 6 to 15, especially 8 to 14, e.g. 10 to 12. However surfactants with HLB as low as 1 may be used.

The surfactant may optionally be or comprise an anionic surfactant such as an ether sulphate, an alkyl benzene sulphonate, an alkyl sulphate, alkane sulphonate, olefin sulphonate, sulposuccinate, sulposuccinamate, soap, sarcosinate, tauride, isethionate, alkyl phosphate, or alkyl ether carboxylate. In each case the surfactant comprises an 8 to 25 carbon alkyl group or alkenyl group or polypropyleneoxy group. Alkyl or alkenyl groups may be straight or branched chain, primary or secondary and preferably have from 10 to 20 eg. 12 to 14 carbon atoms. Ether groups may comprise glyceryl groups and/or 1 to 20 mol polyoxyethylene groups e.g. 2 to 10 mole. The anionic group is usually a sulphate or sulphonate group, but may also be for example a phosphate, phosphonate or carboxylate group.

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The counter ion of the anionic surfactant is usually sodium but may also be potassium, lithium, ammonium or, calcium or other alkali metal or alkaline earth metal.

The surfactant may be or may comprise an amphoteric surfactant such as betaine, sulphobetaine or phosphobetaine. Examples include fatty alkyl dimethyl betaines, alkyl amidopropyl betaines and imidazoline betaines.

The surfactant may, alternatively be or comprise a cationic surfactant such as a C<sub>8-25</sub> straight or branched alkyl or alkenyl or alkylphenyl tri C<sub>1-4</sub> alkyl or hydroxyalkyl ammonium salt, or di C<sub>1-4</sub> alkyl benzyl ammonium salt, or an C<sub>8-20</sub> alkyl or alkenyl amido amine.

#### **ELECTROLYTE**

The presence of electrolyte is not normally required for structuring but is generally tolerated if required for other purposes. We particularly prefer electrolyte-free or low electrolyte (e.g. 1 to 5% by weight) compositions for personal care applications or where clear formulations are required but can tolerate much higher levels, e.g. up to 20% or more if required. For example industrial cleaning formulations may require high levels of alkali such as sodium hydroxide, carbonate or silicate. The presence of builders such as citrate, potassium pyrophosphate, or sodium tripolyphosphate may also be tolerated. Electrolyte may contribute to the structuring of the composition, and may be desirable as a costructurant when very water soluble surfactants or surfactants of high HLB are used.

#### **SUSPENDED MATTER**

The composition may contain suspended solid, liquid or gaseous particles. For instance the composition may contain suspended oil droplets. The oil is preferably a mineral oil (e.g. a low molecular weight petroleum oil) or a fatty glyceride or other

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The structured suspending systems of the invention may be used to suspend builders such as zeolite or sodium tripolyphosphate, agricultural and horticultural pesticides, biocides for water treatment, cuttings or shale in drilling muds, antifoams, explosives, gums such as gum benzoin, guan acacia, gum tragacanth xanthan and guar gum, enzymes, flavouring and vitamin concentrates, calcium phosphate for toothpaste, pharmaceuticals, and machinery and cutting abrasives such as emery or diamond powder.

**PEARLISING**

The compositions of the invention are particularly useful for suspending pearlising agents. Pearlisers are required as concentrates for incorporation into liquid

formulations such as shampoos and toiletries to impart a nacreous iridescence which is attractive to consumers, and can mask inhomogeneities in the formulations.

Pearlisers typically comprise small, thin, transparent platelet crystals which can be suspended in a parallel configuration. When so suspended light falling on the crystals undergoes complex multiple reflections within the substrate similar to those which occur in a pearl and giving rise to similar optical interference effects.

Natural pearls comprise alternate layers of calcium carbonate and protein. Artificial pearlisers which can be suspended according to the invention include guanine/hypoxanthine crystals extracted from fish scales, mica, various salts of lead, zinc, mercury and bismuth (e.g. bismuth oxychloride), titanium oxide and various fatty acid derivatives such as magnesium stearate, coconut monoethanolamide, ethylene glycol distearate and ethylene glycol monostearate. Fish scale extracts are too expensive and the inorganic pearlisers are either too toxic for general use in toiletries e.g. lead, mercury, or relatively ineffective e.g. bismuth. The fatty acid derivatives are therefore now the most widely used pearlisers. In addition to the chemical nature and physical form of the pearliser the manner in which it is suspended has an important effect on its visual impact.

Difficulty is sometimes encountered obtaining the desired effect when incorporating pearlisers into aqueous formulations.

Conventional fatty acid derived pearlisers are supplied as solids which are usually added to a heated formulation above their melting point and recrystallised in situ. The conditions of crystallisation and especially the amount and nature of the agitation applied must be carefully controlled in order to obtain an acceptable result. This makes it difficult to obtain consistent effects and renders solid pearlisers inconvenient to use.

Attempts have been made to prepare liquid concentrates or suspensions which can be added directly to shampoo formulations without heating. While more convenient for

the user. such concentrates face the manufacturer with problems of obtaining a high and consistent pearl effect, similar to those which confront the user of conventional solid pearlisers. Difficulty is also encountered in maintaining the particles in stable suspension and preventing sedimentation.

We have now discovered that carbohydrate structured phases of the invention have the capacity to form stable suspensions of pearlisers.

The pearliser may be dispersed in the aqueous structure surfactant system e.g. by gently stirring, but in the case of the fatty acid derivatives are preferably prepared in situ by heating above their melting point, e.g. temperatures between 65 and 80°C, dispersing the liquid pearliser in the structured surfactant system, preferably with sufficient stirring to form droplets of from 0.5 to 20 microns, e.g. 1 to 10 microns, and cooling to ambient temperature. Preferably cooling is relatively slow e.g. the mixture is allowed to cool naturally. The amount of pearliser can be varied considerably, the main constraint on the upper limit being the viscosity.

The amount of pearliser should not be so high as to render the product unpourable, or unacceptably viscous. We prefer on economic grounds that the pearliser is present in amounts greater than suspending surfactant. Generally pearliser may be present in amounts ranging from 5% up to about 50% e.g. 10 to 45% of the total weight of the mixture.

### **OTHER INGREDIENTS**

The composition may contain minor amounts of other ingredients such as dyes, perfumes, soil suspending agents or optical brighteners. Solvents such as ethanol or isopropyl alcohol ethylene glycol, isopropylene glycol, glycerol or water miscible glycol ethers such as ethylene glycol monomethyl ether, diethylene glycol monomethyl ether or polyethylene glycol, and hydrotypes such as C<sub>1-6</sub> alkyl benzene sulphonates or urea may be required for special applications, e.g. as perfume enhancers, but if not so required are generally undesirable and are preferably absent

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but may be tolerated in small amounts, preferably less than 10%, e.g. less than 5%, most preferably less than 2%.

### OPTICAL VARIEGATION

The present invention is particularly adapted to providing optically variegated fluid surfactant compositions.

Many fluid surfactant - containing products are purchased by the consumer on the basis of factors which include the appearance of the product. Detergents, shampoos, toiletries, soaps and other surfactant-based consumer products often depend upon appearance and packaging for at least part of their consumer appeal. Striped toothpaste, marbled soap and blue speckled detergent powder are well known examples of products whose successful promotion was based on a characteristically variegated appearance.

However it is not, on the face of it, possible to produce any kind of lasting variegation in an otherwise homogenous, pourable, liquid formulation.

It is known to combine two or more immiscible liquids of different density and colour to form a product which segregates into horizontal bands.

The visual effect achievable by this method is limited and the product has the practical disadvantage that the essential functional components are not evenly distributed between the different bands so that the product performs inconsistently if it is not vigorously agitated immediately prior to use.

Structured surfactant s may be used to suspend coloured granules, to produce a speckled effect.



We have now further discovered that where two or more portions of a structured surfactant such as those according to the present invention are separately coloured by including in at least one of said proportions a pigment, which is insoluble in said continuous phase or a dye which is insoluble in said continuous phase and soluble in or absorbable on any dispersed phase and said proportions are charged to a transparent container in such a way as to produce a variegated appearance, little or no migration of pigment or dye through the composition is observed in the undisturbed sample even after prolonged standing. The variegation thus remains stable to a remarkable degree. Furthermore, provided the container is substantially full, even the agitation caused by normal handling during distribution does not significantly affect the variegated appearance of the product. Yet the product may be, to all appearances, a thin, mobile liquid.

According to a further embodiment our invention therefore provides a packaged fluid surfactant-containing product, comprising an at least partially transparent container, and therein a stable structured surfactant comprising a continuous phase and a dispersed phase and having a variegated appearance caused by the inclusion in localised portions of said structured surfactant of a dye or pigment which is insoluble in said continuous phase and present (A) as particles suspended in said continuous phase and having a particle size sufficiently small to be able to give said portions a substantially homogenous appearance which is visually distinct from other portions of said structured surfactant in said container, and/or (B) dissolved in or absorbed on said dispersed phase.

The effects obtainable can be extremely varied. Depending upon how the different visually distinct portions of the structured surfactant are charged to the container it is possible to obtain horizontal or vertical stripes, vertical segments, marbling, bands, whorls or numerous other decorative effects.

Any pattern or optical effect which can be instantaneously obtained by charging visually distinct liquids to a transparent container can be rendered substantially permanent, at least until the product is poured from the container, by using the

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structured surfactants as said liquids. With suitable filling techniques, it is even possible to produce readable characters so that liquid products may be marked with Trademarks, logos or similar devices.

The pigment or dye may for example be, or comprise a water insoluble pigment, having a particle size preferably less than 150 microns especially less than 100 microns, most preferably less than 50 microns e.g. 0.1 to 20 microns.

The proportion of pigment required is generally small. Any pigmented portion normally only requires from 0.001 to 1% by weight of pigment to produce a sufficient effect e.g. 0.01 to 0.5% more usually 0.02 to 0.1%. The precise amount will depend on the choice of pigment and the intensity of colour required.

The pigment may be a white pigment (e.g. titanium dioxide) a black pigment (e.g. carbon black), coloured pigment such as any water insoluble pigment hitherto used in cosmetics or detergents, or a pearlescing agent such as mica.

Typically the structured surfactant has an aqueous continuous phase and the pigment or dye is water insoluble. Water insoluble dyes typically dissolve in or are absorbed on a dispersed surfactant phase.

We prefer that, apart from the pigment or dye, the differently coloured portions of the structured surfactant product of our invention should have essentially the same composition. This assists maintaining the stability of the product and ensures uniform performance. However it is possible e.g. where substantially water insoluble active ingredients other than pigment are suspended in the product or dissolved or otherwise contained in a suspended oil phase, to include such ingredients in only some portions of the product. This may be useful in segregating mutually incompatible components and may permit promotional claims that the coloured portions are associated with a specific beneficial effect.

The method of filling the container determines the effects produced. For example horizontal stripes can be obtained by running a single bladed stirrer gently in a container partly filled with one coloured structured surfactant and injecting a contrasting colour through a syringe at the level of the stirrer. Progressively raising the stirrer and repeating the process produces a plurality of horizontal stripes. Injecting the contrasting liquid while drawing the syringe up the side of the container produces a vertical stripe. Inserting a partition into the container, and filling different portions on either side, before withdrawing the partition, produces contrasting vertical halves. A multiple partition permits vertical segments. Alternatively vertical effects may be produced by inserting two or more tubes into the container and gradually withdrawing the tubes while charging the container with different coloured portions, each at the same rate.

Partially stirring two contrasting portions together before filling the container gives an attractive marbled effect. The foregoing are merely an indication of some of the different filling techniques and associated visual effects possible according to the invention. Numerous other possibilities will be apparent to those skilled in the art.

The container may be any jar, bottle, tube, sachet or other conventional container for surfactant based products. It may typically be of glass or plastic or other transparent material. It may be coloured but is preferably at least partly clear to enable the decorative contents to be easily seen. It is possible to use deformable containers such as squeeze tubes or sachets, provided that they are sufficiently filled to give them a degree of rigidity enough to avoid loss of variegation on normal handling prior to use, but it is preferred to use rigid or at least substantially non-deformable materials.

The invention will be illustrated by the following examples in which all proportions are based on weight percentages of active ingredient based on the total weight of the composition, unless stated to the contrary.

**Examples 1 to 4**

The following formulations were prepared:-

	1	2	3	4
C <sub>12-14</sub> 3 mole ether sulphate	10	12.5	15	17.5
Sucrose	46	46	46	46
Trisodium citrate	2	2	2	2
Perfume	5	5	5	5
Water	balance	balance	balance	balance

All four formulations were clear or slightly hazy, mobile structured liquids with good suspending properties. Suspensions of talc, mineral oil, pigment, small beads and plastic novelty items were prepared. All were stable after prolonged storage.

Each of the examples 1 to 4 was re-prepared (a) without the perfume (b) without the citrate and (c) without perfume or citrate. No suspending power was exhibited by any of the eight samples of preparations (a) and (c). The samples of preparation (b) all exhibited similar suspending power to the original examples. On heating the samples to 70°C and subsequent cooling a clear transparent composition was obtained.

A clear sample of Example 1(b) and a sample containing a red pigment in suspension were slowly poured into a sample jar in a series of alternating additions. The effect was to produce a sequence of horizontal stripes. When the bottle was filled the stripes retained their integrity and showed no signs of blurring or diffusion after one year storage including intermittent periods of gentle shaking and six months weeks stored on its side.

**Example 5**

A schematic phase diagram was prepared for the system coconut diethanolamide/sucrose/water and is reproduced as fig. 1 of the drawings. The area marked "clear lamellar" represents transparent expanded G-phases having a

suspending power according to the invention, and the area marked "lamellar" comprises opaque expanded lamellar suspending systems according to the invention. The phase boundaries illustrated were not all precisely determined.

#### **Example 6**

A sample was prepared comprising 10% coconut diethanolamide, 35% water and 55% sucrose. The product was a clear attenuated G-phase with good suspending power.

#### **Example 7**

A composition comprising 10% C<sub>12-14</sub> alkyl 2 mole ethoxysulphate, 33% water, 50% sucrose, 5% ethanol based perfume, 2% sodium citrate gave a clear, attenuated G-phase with good suspending powers.

#### **Example 8 (Shampoo formulation)**

4 parts by weight of the composition of Example 7 and 1 part of the composition of Example 6 were mixed together to form a clear composition of the invention with good suspending powers and good performance as a skin cleanser and shampoo.

#### **Example 9 (laundry detergent formulation)**

	% Active ingredient by weight
C <sub>12-14</sub> linear alkyl benzene sulphonate	6.6
Triethanolamine lauryl sulphate	1.65
C <sub>12-14</sub> alkyl 3 mole ethoxylate	1.6
Sucrose	55.0
Sodium diethlenetriamine pentakis (methylenephosphonate)	0.55
Water	balance

The product was a hazy, readily pourable liquid with good suspending power.

#### **Example 10 (Pearl concentrate)**

A pearl concentrate was obtained by heating a formulation comprising 54% by weight sucrose, 10% by weight coconut di-ethanolamide, 10% by weight ethylene glycol distearate and 26% by weight water to 70°C and cooling.

A spontaneously pearly suspension was obtained.

#### **Example 11**

10% by weight C<sub>12-14</sub> alkyl six mole ethoxylate (HLB = 10), 54% by weight sucrose and 36% by weight water, were mixed and warmed to 70°C.

The cooled product was a clear transparent, pourable system with good suspending properties.

#### **Example 12**

	% w/w
sucrose	40.0
perfume	2.0
sodium C <sub>12-14</sub> alkyl 3 mole ethoxy sulphate (70%)	8.0
coconut diethanolamide	2.0
sodium chloride	5.0
water	balance

The above formulation provided a clear, transparent, pourable fluid. Whorls of three different coloured pigments were introduced into this formulation with a syringe. After three months no diffusion of the pigment was observable.

**Example 13**

	% w/w
sucrose	40.0
perfume	2.0
trisodium citrate dihydrate	2.0
sodium C <sub>12-14</sub> alkyl 3 mole ethoxy sulphate (70% by wt active)	8.8
glycamate	5.06
coconut monoethanolamide	1.1
coconut amido propyl betaine	4.18
sodium C <sub>8-10</sub> alkyl polyglycoside dp 1.6 (65%)	6.2
sodium chloride	0.66
sodium ethylene diamine tetraacetate	0.05
water	balance

The composition was a pourable, clear, transparent fluid with suspending properties.

A plurality of coloured, polystyrene beads (1mm diameter) were dispersed in the composition. The suspension remained stable after three months.

**Example 14**

	% w/w
sodium C <sub>12-14</sub> alkyl (3 mole ethoxy sulphate)	10.0
coconut diethanolamide	2.5
fructose	50.0
water	balance

The above composition was a clear isotropic L<sub>1</sub> micellar solution which was unsaturated and had no suspending power. Addition of 6% by weight sodium chloride, gave a stable, easily pourable fluid, composition which after shaking was capable of suspending air bubbles. The aerated composition was stood overnight at 50°C. The aged composition was clear and transparent and maintained the air bubbles in a stable suspension. Equivalent compositions with 2 and 4% respectively of sodium chloride were not able to suspend bubbles under the foregoing conditions.

**Example 15**

	% w/w
sodium C <sub>12-14</sub> alkyl -3 mole ethoxy sulphate	8.0 A.I.
coconut diethanolamide + 10% by wt glycerol	2.0 A.I.
fructose	40.0
water	balance

The above composition was not capable of maintaining particles in suspension.

Addition of incremental amounts of sodium chloride gave the following results:-

**wt % sodium chloride**

2	L1 phase bubbles rise
4	L1 phase bubbles rise
6	clear suspending phase. Bubbles suspended after ageing at 50°C overnight.

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CLAIMS

1. The use of water soluble surfactants as structurants in structured surfactant systems.
2. A structured surfactant system having suspending properties which comprises a surfactant, water and a structurant characterised in that said structurant comprises a water soluble carbohydrate.
3. A system according to claim 2 which consists of a G-phase.
4. A system according to claim 3 wherein said G-phase has a lamellar repeat spacing greater than 15nm.
5. A composition according to claim 4 which is transparent.
6. A structured surfactant system having suspending properties and surfactant, carbohydrate and water and having a structural feature with a repeat spacing of from 20 to 50nm.
7. A system according to claim 4 wherein said feature is lamellar.
8. A system according to any of claims 2 to 7 wherein said carbohydrate is present in any amount of at least 25% by weight of the mixture of water surfactant carbohydrate.
9. A system according to any of claims 2 to 8 wherein the surfactant is present in an amount of from 1 to 60% by weight of the system.
10. A system according to any of claims 2 to 9 containing more than 20% by weight water.

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11. A system according to claim 10 containing 30 to 60% by weight water.
12. A composition according to any of claims 2 to 11 wherein the carbohydrate is a mono or disaccharide sugar, gluconic acid, mannlic acid, ascorbic acid, sorbitol, mannitol or inositol.
13. An aqueous suspension comprising a system according to any of claims 2 to 7 and particles of solid, liquid or gas stably suspended therein.
14. A composition according to claim 13 wherein said particles comprise a builder.
15. A composition according to either of claims 13 and 14 wherein said particles comprise an abrasive.
16. A composition according to any of claims 13 to 16 wherein said particles comprise a pesticide.
17. A composition according to any of claims 13 to 16 wherein said particles comprise an oil.
18. A composition according to any of claims 13 to 17 wherein said particles comprise a pigment.
19. A composition according to claim 18 having a plurality of differently pigmented zones producing a variegated visual effect.
20. A packaged fluid surfactant-containing product, comprising an at least partially transparent container, and therein a stable structured surfactant comprising a continuous phase and a dispersed phase and having a variegated appearance caused by the inclusion in localised portions of said structured surfactant of a dye or pigment which is insoluble in said continuous phase and

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present (A) as particles suspended in said continuous phase and having a particle size sufficiently small to be able to give said portions a substantially homogenous appearance which is visually distinct from other portions of said structured surfactant in said container, and/or (B) dissolved in or absorbed on said dispersed phase.

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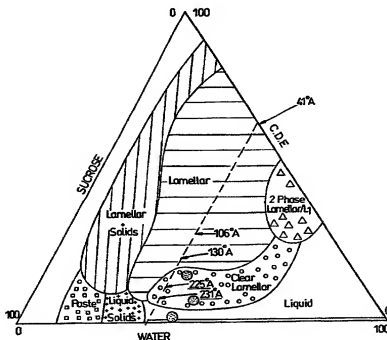
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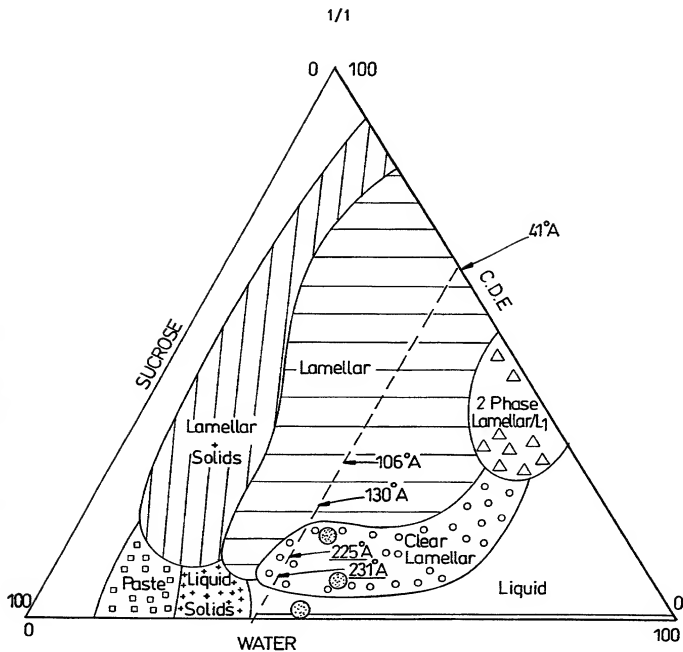
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(57) Abstract: Carbohydrates are used to confer structure on a structured surfactant system comprising surfactant and water and capable of suspending particles of solid, liquid and/or gas.

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# HUNTSMAN

## Power of Attorney

I, Peter R. Huntsman, hereby certify that I am an officer of Huntsman International LLC. (the "Corporation"), a corporation, duly organized, validly existing and in good standing under the laws of the State of Delaware, in the United States of America, with the authority to and hereby grant, on behalf of the Corporation:

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Marnix Karel Christiane Moens,  
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- (i) appoint attorneys or agents in any and all jurisdictions, including other persons within the Corporation, and delegate to or revoke from them the same, or any portion thereof, powers hereinbefore conferred as deemed necessary or advisable for the promotion of the Purpose;
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- (iii) generally to sign, execute, give and accept all such papers, communications and other items as may be necessary or advisable for the promotion of the Purpose.

IN WITNESS WHEREOF, the Corporation have executed this Certificate and Power to be effective as of January 1<sup>st</sup>, 2001.

Huntsman International LLC.

By 

Date 01 JAN. 2001

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**DECLARATION FOR PATENT APPLICATION**

As an undersigned inventor, I hereby declare that:

My residence, post office address and country of citizenship are as stated directly below my name.

I believe (check one) ☒ I am the original, first and sole inventor  
☐ I am a joint inventor and the below named inventors are the original and first inventors

of the subject matter which is claimed and for which a patent is sought on the invention entitled **"STRUCTURED SURFACTANT SYSTEMS"** (Attorney Docket **MPL318**), the specification of which

(check one) ☐ is attached hereto.

☒ was filed on July 14, 2000  
as Application Serial No. PCT/GB00/02725  
and was amended on \_\_\_\_\_  
(if applicable)

I further declare that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose to the United States Patent and Trademark Office (hereinafter "the Office") all information known to me to be material to patentability of the subject matter which is claimed as defined in 37 C.F.R. §1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119 of any foreign application(s) for patent or inventor's certificate indicated below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s) Number	Country	Day/Month/Year Filed	Priority Claimed	
			Yes	No
<u>9916744.7</u>	<u>Great Britain</u>	<u>17 July 1999</u>	<input checked="" type="checkbox"/>	<input type="checkbox"/>
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<u>9925700.8</u>	<u>Great Britain</u>	<u>30 October 1999</u>	<input checked="" type="checkbox"/>	<input type="checkbox"/>
<u>9927768.3</u>	<u>Great Britain</u>	<u>25 November 1999</u>	<input checked="" type="checkbox"/>	<input type="checkbox"/>

I hereby claim the benefit under 35 U.S.C. §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of 35 U.S.C. §112,

I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in 37 C.F.R. §1.56, which became available between the filing date of the prior application and the national or PCT international filing date of this application: None.

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that these statements were made with the knowledge that willful false statements and the like are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and may jeopardize the validity of the application or any patent issued thereon.

Full name of inventor John Hawkins

Inventor's Signature [Signature] 31-1-2002

Date

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